Supplementary Information

Negligible effects from sample preparation

No OH bands have been observed in the dehydrated grains of An_{68-70}, An_{96}, and GRR 2058 using FTIR. Furthermore, the unheated An_{68-70} and An_{96} grains contain 10 and 7 ppm H_2O, respectively (Table 1), while the dehydrated grains of An_{68-70}, An_{96}, and GRR 2058 all display uniform $OH^-/^{30}Si^-$ values, lower than that of the unheated GRR 2058 (~1 ppm) (Fig. 2). Therefore, these dehydrated grains have lost their original OH contents, any hydrogen detected in the dehydrated grains is assumed to be background in the SIMS measurements, and sample preparation has not introduced additional hydrogen signal.

Hydrogen isotope fractionation between mineral and melt

No experimental studies have been carried out on the fractionation of hydrogen isotope between mineral and melt. The fractionation during mantle melting and magma differentiation, however, have been discussed using hydrogen isotope ratios of fresh submarine basalts (Kyser and O'Neil, 1984; Bindeman et al., 2012). It has been suggested differentiation or partial melting have had little effect on the hydrogen isotope fractionation (Kyser and O'Neil, 1984; Bindeman et al., 2012). Melt-lherzolite fractionation of hydrogen isotope has been suggested to be less than 10‰ during mantle melting (Bindeman et al., 2012). The uncertainties of δD of undegassed lunar minerals are rather large, typically >100‰ (Barnes et al., 2014). Therefore, the δD of lunar plutonic minerals could represent those of their parent magmas.

The δD value of GRR1968

The low water content (70 ppm) of GRR 1968 has made the determination of its hydrogen isotope ratio using traditional method (e.g., mass spectrometer) on bulk sample difficult. In this study, we use an alternative approach to calculate the δD value of GRR1968.

GRR1968 is an anorthite ($An_{93.8}Ab_{4.0}$ with 0.37% of FeO) megacryst of island arc basalt from Miyake Island of Izu-Bonin-Mariana island arc (Kimata et al., 1995; Johnson and Rossman, 2003). Because the fractionation of hydrogen isotope between mineral and melt is very small (see text above), we could use the melt δD value to represent that of GRR1968. No hydrogen isotope ratios have been published for arc basalts from Miyake Island. Therefore, we use the average δD value (-42 ± 29‰) of samples from Izu-Bonin-Mariana arc system found in the literature. This dataset includes 87 samples with δD varying from -12 to -73.7‰ (Table S4, and references therein). Furthermore, it has been suggested that the fractionation of hydrogen isotope between mineral and melt is less than 10‰ (Kyser and O'Neil, 1984; Bindeman et al., 2012). Therefore, we estimate that the δD value of GRR1968 is -42 ± 39‰. This δD estimation is reasonable and sufficiently precise for the purpose of our study, because most of the uncertainties in the H isotopic results of lunar plagioclases are from the correction for cosmogenic production of hydrogen. Furthermore, the δD of An_{96}, a plagioclase from Aleutian arc melt (Waythomas et al., 2010), is -10 ± 90‰ using this standard GRR1968 (-42 ± 39‰), overlapping with those (-12 to -73.7‰) from Izu-Bonin-Mariana arc system (Table S4, and references therein).

Estimation of the initial LMO water content

The plagioclase of ferroan anorthosite 60015 contains 5 ± 1 ppm water analyzed by SIMS. Using a plagioclase-melt partition coefficient (Hamada et al., 2013; 0.005 ± 0.003 if the

latest infrared absorption coefficient determined for water in plagioclase by Mosenfelder et al. (2015) is used), the LMO melt that equilibrated with the ferroan anorthositic plagioclase could have contained 1000 ppm water. The relatively large uncertainty primarily comes from that of partition coefficient. Water in the LMO could be lost through degassing into a vacuum. On the other hand, water in the LMO melt could increase as the LMO solidification continued. If only these two scenarios were considered, we can infer the initial LMO water content.

The hydrogen species degassed in the Moon has been suggested to be H_2 (Sharp et al., 2013). The fraction of hydrogen (equivalent to water) lost from the LMO into a vacuum before the crystallization of ferroan anorthosite can be estimated using the Rayleigh fractionation equation $\frac{R}{R_0} = F^{\sqrt{M_1/M_2}-1}$, where R is the D/H ratio of the LMO when a fraction F of hydrogen remains in the LMO, and R_0 is the initial hydrogen isotope ratio of the LMO, and M_1 and M_2 are the masses of the volatile phase isotopologues (masses of 2.016 for H_2, and 3.022 for HD; Sharp et al., 2013). The δD of -281 ± 49‰ (represented by sample 77215) is used as the initial hydrogen isotope ratio of the LMO, whereas +310 ± 110‰ (represented by sample 60015) is used as the final hydrogen isotope ratio of the LMO. Therefore, the fraction of hydrogen remained in the LMO when 60015 crystallized is 3.8%. Furthermore, 20 ± 5% of the LMO may have remained melt when ferroan anorthositic plagioclase crystallized (Shearer et al., 2006; Elkins-Tanton et al., 2011). Note almost all of the water undegassed from the LMO (i.e., 3.8% of initial LMO water) should have still remained in the LMO melt residue. Therefore, the initial LMO water content could have been up to 5000 $\pm^{6600}_{2900}$ ppm. If a new partition coefficient of 0.02 ± 0.002 determined under the lunar condition by Caseres et al. (2017), which is also in the range of 0.006 – 0.04 determined by Lin et al. (2017), is used, the calculated initial LMO water content would be 1320 $\pm^{1240}_{680}$ppm. However, if a generic partition coefficient of 0.001 used by Elkins-Tanton and Grove (2011) in their LMO model is also used here, the initial LMO water content would be ~2.6 wt%. Therefore, an accurate hydrogen partition coefficient between plagioclase and lunar melt is needed to model the degassing process in the LMO.

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